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DETAILED DESCRIPTION

[Detailed Description of the Invention] [0001]

[Field of the Invention] This invention relates to the cosmetics, ultraviolet inhibitor, and foodstuffs in which the anti-oxidant and anti-oxidant which control prolonged oxidation to the cosmetics by which the anti-oxidant and the anti-oxidant were added, ultraviolet inhibitor and foodstuffs, especially lipid were added.

[0002]

[Description of the Prior Art]In recent years, antioxidizing of sebum made into the cause of aging of skin has attracted attention. And research is advanced as an anti-oxidant of this sebum, for example about tocopherol (it is also called vitamin E) or ascorbic acid (it is also called vitamin C).

[0003]For example, in the experiment using the swine skin, Douglas Darr and others has reported that vitamin C is effective as a synergistic protective agent when UVB sun screen use is carried out to the damage (sunburn cell formation) by an acute ultraviolet-rays B wave (UVB). It is reported that the dramatically outstanding defense effect was acquired from the UVB obstacle by concomitant use of vitamin E and vitamin C. moreover -- vitamin C excels vitamin E in the point defended from the phototoxicity damage by an ultravioletrays A wave (UVA) clearly -- the thing of concomitant use of vitamin C and vitamin E -vitamin C -- it is reported that it was only more slightly [independently] effective. And if it blends together with vitamin C or vitamin C, and the UVA/B sun block (for example, oxybenzone) of marketing of both vitamin E, As opposed to phototoxic damage. . There is a report that the clearly high defense effect was accepted. (Stanley) [Douglas Darr,] Dunston and Holly Faust, Sheldon Pinnell. Effective. of. 76 Antioxidants (Vitamin C and E) With and Without Sunscreens as Topical Photoprotectants. Acta Derm Venereol(s) (stockh), No. 4. 264 - 268 pages, and 1996. Here, ultraviolet rays (maximal absorption wavelength of 290 nm) are roughly divided into two, and are divided into an ultraviolet-rays B wave (UVB) and an ultraviolet-rays A wave (UVA). An ultraviolet-rays B wave (UVB) points out the ultraviolet rays of nearby long wavelength in ultraviolet rays, and says the

ultraviolet rays which reach the depths of the skin. An ultraviolet-rays A wave (UVA) points out the ultraviolet rays (320-400 nm) of nearby short wavelength in ultraviolet rays, and says the ultraviolet rays which reach only to the surface portion of the skin.

[0004]Generally generation of peroxylipid in the living body is divided roughly into the system which carried out conjugate to the enzyme reaction under metal ion existence, and the nonenzymatic reaction derived with a metal ion, ascorbic acid, etc. In the former, the intervention of a hydroxy radical (-OH), active oxygen like singlet oxygen, or an iron-oxygen complex is suggested.

[0005]Samura and others by what (vitamin E does not oxidize) vitamin E supplements with the free radical generated in process of these active oxygen and lipid peroxidation at this time, or active oxygen and an iron-oxygen complex are inactivated for. It reported that an antioxidant action was shown, and also if reducing agents, such as sufficient vitamin C, exist when vitamin E oxidizes, it will have reported that it is returned and a vitamin-E radical reproduces vitamin E. In [it is reported that vitamin E has an optical protective action, and] a manifestation and development of the wrinkles of the skin surface after the photoaging (photoaging), i.e., an ultraviolet-rays (especially UVB) exposure, Vitamin E delays the manifestation of wrinkles and controlling development of wrinkles is reported (Samura 1 ***, the Morita ***, "the function in the skin of vitamin E and application", FRAGRANCE JOURNAL special issue No.15 (1996), 169 - 178 pages).

[0006]It is pointed out to the cardiopathy by arteriosclerosis in recent years that not the lipid itself but the lipid which carried out oxidation denaturation has an important problem.

[0007]

[Problem(s) to be Solved by the Invention] However, if extensive administration of the vitamin E is carried out, and antioxidizing of lipid will also have the report that peroxylipid increases conversely and will only be medicated with vitamin E, the antioxidant action to peroxylipid in the living body will not necessarily be obtained.

[0008]If vitamin C and vitamin E are used together, even if it is reported that an antioxidant action occurs, the optimum percentage is not reported at all.

[0009]This invention is made in order to solve the aforementioned technical problem, and it is a thing.

By making an anti-oxidant contain the dimer of **, the purpose is to provide lasting high anti-oxidants and these cosmetics to contain, and ultraviolet inhibitor.

[0010]

[Means for Solving the Problem]In order to solve the above-mentioned technical problem, an anti-oxidant concerning this invention has the following features.

[0011](1) An anti-oxidant contains a dimer of tocopherol.

[0012]Generally, lipid is made radical, a lipid radical generates, and also a lipid radical oxidizes and generates a lipid-peroxidation radical. Tocopherol has the operation which makes generation of this lipid-peroxidation radical control.

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[0019]By adding tocopherol in composition of an anti-oxidant of the above (3), an antioxidant action increases further.

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[0021]By adding a lipophilic group salt of ascorbic acid with the above-mentioned addition, it will have the optimal antioxidant action. Even if it adds more, an antioxidant action does not increase.

[0022](6) They are cosmetics which contain an anti-oxidant of a statement in either of (5) from the above (1).

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[0025]

[Embodiment of the Invention]Hereafter, an embodiment of the invention is described using figures.

[0026]Conventional tocopherol returned the oxide in the oxidation mechanism above the dotted line of <u>drawing 1</u>, it became a tocopherol radical, and it was thought that an antioxidant action was shown. However, invention-in-this-application persons discovered the antioxidant action mechanism of tocopherol as shown in the dotted-line bottom of drawing 1.

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peroxidation radical (ROO-) here, Returning the lipid-peroxidation radical to hydronalium peroxide (ROOH), by becoming alpha-TOKOFEROKISHIRAJIKARU (A-), self controlled lipid radical-ization and has controlled oxidation of lipid as a result (Step 6). The reaction of this antioxidation is an antioxidant action mechanism of the tocopherol known conventionally.

[0028]The mechanism of action shown below is clarified by invention-in-this-application persons.

[0029]alpha-TOKOFEROKISHIRAJIKARU (A-) generated by returning a lipid-peroxidation radical (ROO-) becomes an alpha-tocopherol dimer (A_2) (Step 7). This alpha-Toco phenol dimer (A_2) reacts to a lipid radical (R-), becomes a stable substance called the stable substance (RA₂) which is a lipid tocopherol dimer substance, and controls oxidation of lipid (Step 8).

[0030]As an alpha-tocopherol dimer is shown in <u>drawing 2</u>, it is separated by high performance chromatography (HPLC), a molecular weight is identified by LC-mass, and it is checked that it is a dimer of tocopherol.

[0031]As shown in <u>drawing 3</u>, while the alpha-tocopherol was consumed, the dimer of the alpha-tocopherol increased (the 2nd step of <u>drawing 3</u>), the dimers of the alpha-tocopherol decreased in number, and while disappearing, it became clear that precipitative oxidation of lipid was started (the 3rd step of <u>drawing 3</u>).

[0032]usually -- as shown in <u>drawing 4</u>, carry out the whole-quantity pair of the oxidation action of linolic acid which is the lipid only by the alpha-tocopherol, and, less than [0.01wt%], it has the antioxidation effect from the first stage -- on the other hand -- more than 0.01wt% -- oxidation being promoted in early stages, if it adds, but. The action that oxidation is controlled after that is shown. In particular, in the case of an alpha-tocopherol simple substance, the balance of oxidation depressant action has the best time of 0.01wt%.

[0033]On the other hand, as shown in <u>drawing 5</u>, ascorbic acid (it omits the following "VC-IP"), for example, oleophilic ascorbic acid tetraisopulmitic acid ester, shows acid oxidation depressor effect from the first stage.

[0034]However, as shown in <u>drawing 4</u>, 5 hours after setting to the temperature 333K, oxidation of alpha-tocopherol 0.01wt% of case starts. However, when 10K temperature goes up, since this oxidation time generally speeds up for 2.5 hours, in the case of the temperature 343K, it will be called 2.5 hours. on the other hand -- VC-IP -- a maximum of 20 wt(s)% -- even if it adds, oxidation is started in 2 hours.

[0035]Therefore, if the oxidation volume effect of an above-mentioned simple substance is taken into consideration and both will usually be added when VC-IP is added to alphatocopherol 0.01wt%, at the temperature 343K, it will be guessed that oxidation depressant action continues only for 4.5 hours.

[0036]However, according to the invention in this application, when VC-IP was added to

alpha-tocopherol 0.01wt%, oxidation depressant action actually continued at the temperature 343K for as long as 14 hours.

[0037]As mentioned above, when the invention in this application contained the dimer of the alpha-tocopherol, the oxidation depressant action beyond the combination of the alphatocopherol and VC-IP was revealed.

[0038]That is, since the lipid shown in <u>drawing 1</u> controls the oxidation of a lipid radical made radical, the dimer of the alpha-tocopherol can aim at oxidation control of the further lipid compared with combination with the alpha-tocopherol and the alpha-tocopherol, ascorbic acid, or its derivative.

[0039]As lipid of the object of the antioxidant action in this embodiment, fatty acid of the straight chain of the carbon numbers 12-20 is mentioned, for example, and the squalene etc. which are contained in linolic acid or sebum are contained.

[0040]As a derivative of the above-mentioned ascorbic acid, For example, sodium ascorbate, ascorbic acid magnesium, phosphorylation vitamin C, The vitamin C which has the hydrophilic nature and lipophilic property which fatty acid ester, grape sugar, and fatty acid of ascorbic acid, such as an ascorbic acid SHITEA rate, ascorbyl palmitate, and ascorbic acid tetraiso palmitate, combined is mentioned. This example explains taking the case of ascorbic acid tetraiso PAMITETO.

[0041]As shown in <u>drawing 3</u>, by making the addition of ascorbic acid tetraiso palmitate increase shows that an antioxidant action increases. In <u>drawing 3</u>, the antioxidant action in 343K is measured linolic acid 100wt%.

[0042]The cosmetics of this embodiment are cosmetics which the above-mentioned antioxidant contains.

[0043]As cosmetics, it is preferred to add for both basic cosmetics and the cosmetics for a makeup.

[0044]Titanium oxide (TiO₂), a zinc oxide, etc. are used for ultraviolet inhibitor, especially the sun block of cosmetics. However, it functions on the above-mentioned titanium oxide also as a photocatalyst, and participating in oxidation of sebum is pointed out. [0045]Ultraviolet inhibitor of this embodiment contains the above-mentioned anti-oxidant. [0046]Therefore, if the anti-oxidant of this embodiment is contained even if titanium oxide

[0046]Therefore, if the anti-oxidant of this embodiment is contained even if titanium oxide and the zinc oxide in ultraviolet inhibitor serve as a photocatalyst and oxidize sebum by ultraviolet rays, the oxidation on and after the early stages of lipid oxidation reaction and the middle can be controlled.

[0047]

[Example]The outline of the experiment measure in which it used for this example is shown in <experimental device and experimental method> drawing 6. The reaction vessel 10 is a three bubbling tower type cylinder mouth glass tube with a content volume of about 200 ml, and is inserted in the thermostat adjusted to prescribed temperature. Into the reaction vessel 10, insertion immobilization of the thermometer 12 which measures the temperature in a reaction vessel is carried out. The opening 18 which can pour in and can extract a

reaction solution is formed. The three-way cock 16 is installed in gas output port so that a gas exchange is possible.

[0048]First, about 40g of linolic acid which is a reaction solution and alpha-tocopherol of the specified quantity, Or the concomitant use solution of the alpha-tocopherol and the ascorbyl palmitate was taught to the reaction vessel 10 from the opening 18, and nitrogen gas was fed by about 300 ml/min from the gas loading slot 14 of the lower part of the reaction vessel 10. The temperature of the thermostat was raised and the temperature of the reaction solution in the reaction vessel 10 was raised to a predetermined temperature. When the temperature of the reaction solution turned into prescribed temperature, nitrogen gas was changed to oxygen and the nitrogen gas of prescribed concentration, and it let the glass filter 15 pass, and with the constant flow of 300 ml/min, it was made to feed in a reaction solution and oxidized. Under [a fixed quantity / peroxide concentration / iodometric titration flow method / it extracts a reaction solution from the opening 18 for every time of a reaction start, and fixed time after a reaction start and].

[0049]Since a little peroxides currently mixed in linolic acid before a reaction start affect the oxidation reaction rate of linolic acid, linolic acid, The Tokyo Chemicals reagent first class goods were heated for about 15 minutes under 365K and a heavy nitrogen air current, the peroxide in linolic acid was disassembled, the concentration was made into 0.5 or less meq/kg, and the thing after decomposition was made into the above-mentioned linolic acid sample for an experiment.

[0050]In this example, what was supplied from Riken Vitamin Co., Ltd. was used as alphatocopherol. As ascorbyl palmitate, what was supplied from Tokyo Chemicals, Inc. was used.

[0051]The example 1. reaction temperature 333K and the concentration of the alphatocopherol were fixed to 1.0 weight sections, and the antioxidant action was considered by changing ascorbic acid tetraiso PARIMITETO concentration. It carried out based on the experimental method. It carried out based on the experimental method. A result is shown in drawing 3.

[0052]The antioxidant action also increased as were shown in drawing 3 and ascorbic acid tetraiso PARIMITETO concentration became high.

[0054]As shown in drawing 7, from the oxygen cylinder 1 and the nitrogen cylinder 2, respectively oxygen gas and nitrogen gas, Needle valvea [3] and 3b and flow-meter 4a.4b, the three-way cocks 5a and 5b, And the three-way cock 5c is supplied via the drying pipes 6a and 6b containing silica gel, and the drying pipes 7a and 7b containing a calcium chloride as a drying pipe, in the three-way cock 5c, distributed gas is exchanged if needed

and the bubble column reaction vessel 10 is supplied. The reaction vessel 10 is allocated in the oil bath 9, and also these are installed in the dark room 11. In the dark room 11, the ultraviolet ray lamp 13 which can irradiate the reaction vessel 10 with ultraviolet rays is formed.

[0055]Under UV irradiation, even if titanium oxide existed in the system, the oxidation rate in early stages of oxidation reaction was delayed, and it became clear that an antioxidant action increased.

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PRIOR ART

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TECHNICAL PROBLEM

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MEANS

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[0025]

[Embodiment of the Invention]Hereafter, an embodiment of the invention is described using figures.

[0026]Conventional tocopherol returned the oxide in the oxidation mechanism above the dotted line of <u>drawing 1</u>, it became a tocopherol radical, and it was thought that an antioxidant action was shown. However, invention-in-this-application persons discovered the antioxidant action mechanism of tocopherol as shown in the dotted-line bottom of drawing 1.

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[0.01wt%], it has the antioxidation effect from the first stage -- on the other hand -- more than 0.01wt% -- oxidation being promoted in early stages, if it adds, but. The action that oxidation is controlled after that is shown. In particular, in the case of an alpha-tocopherol simple substance, the balance of oxidation depressant action has the best time of 0.01wt%.

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EXAMPLE

[Example]The outline of the experiment measure in which it used for this example is shown in <experimental device and experimental method> drawing 6. The reaction vessel 10 is a three bubbling tower type cylinder mouth glass tube with a content volume of about 200 ml, and is inserted in the thermostat adjusted to prescribed temperature. Into the reaction vessel 10, insertion immobilization of the thermometer 12 which measures the temperature in a reaction vessel is carried out. The opening 18 which can pour in and can extract a reaction solution is formed. The three-way cock 16 is installed in gas output port so that a gas exchange is possible.

[0048]First, about 40g of linolic acid which is a reaction solution and alpha-tocopherol of the specified quantity, Or the concomitant use solution of the alpha-tocopherol and the ascorbyl palmitate was taught to the reaction vessel 10 from the opening 18, and nitrogen gas was fed by about 300 ml/min from the gas loading slot 14 of the lower part of the reaction vessel 10. The temperature of the thermostat was raised and the temperature of the reaction solution in the reaction vessel 10 was raised to a predetermined temperature. When the temperature of the reaction solution turned into prescribed temperature, nitrogen gas was changed to oxygen and the nitrogen gas of prescribed concentration, and it let the glass filter 15 pass, and with the constant flow of 300 ml/min, it was made to feed in a reaction solution and oxidized. Under [a fixed quantity / peroxide concentration / iodometric titration flow method / it extracts a reaction solution from the opening 18 for every time of a reaction start, and fixed time after a reaction start and].

[0049]Since a little peroxides currently mixed in linolic acid before a reaction start affect the oxidation reaction rate of linolic acid, linolic acid, The Tokyo Chemicals reagent first class goods were heated for about 15 minutes under 365K and a heavy nitrogen air current, the peroxide in linolic acid was disassembled, the concentration was made into 0.5 or less meq/kg, and the thing after decomposition was made into the above-mentioned linolic acid sample for an experiment.

[0050]In this example, what was supplied from Riken Vitamin Co., Ltd. was used as alphatocopherol. As ascorbyl palmitate, what was supplied from Tokyo Chemicals, Inc. was

used.

[0051]The example 1. reaction temperature 333K and the concentration of the alphatocopherol were fixed to 1.0 weight sections, and the antioxidant action was considered by changing ascorbic acid tetraiso PARIMITETO concentration. It carried out based on the experimental method. It carried out based on the experimental method. A result is shown in drawing 3.

[0052]The antioxidant action also increased as were shown in drawing 3 and ascorbic acid tetraiso PARIMITETO concentration became high.

[0054]As shown in drawing 7, from the oxygen cylinder 1 and the nitrogen cylinder 2, respectively oxygen gas and nitrogen gas, Needle valvea [3] and 3b and flow-meter 4a.4b, the three-way cocks 5a and 5b, And the three-way cock 5c is supplied via the drying pipes 6a and 6b containing silica gel, and the drying pipes 7a and 7b containing a calcium chloride as a drying pipe, in the three-way cock 5c, distributed gas is exchanged if needed and the bubble column reaction vessel 10 is supplied. The reaction vessel 10 is allocated in the oil bath 9, and also these are installed in the dark room 11. In the dark room 11, the ultraviolet ray lamp 13 which can irradiate the reaction vessel 10 with ultraviolet rays is formed.

[0055]Under UV irradiation, even if titanium oxide existed in the system, the oxidation rate in early stages of oxidation reaction was delayed, and it became clear that an antioxidant action increased.

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1]It is a figure explaining the mechanism of the lipid antioxidant action of the alpha-tocopherol.

[Drawing 2]It is a figure explaining identification of the alpha-tocopherol dimer generated from the alpha-tocopherol at the time of an antioxidant action.

[Drawing 3]It is a figure explaining the influence of an antioxidant action when alphatocopherol concentration is fixed and ascorbic acid tetraiso palmitate concentration is changed.

[Drawing 4] It is a figure explaining the addition of the alpha-tocopherol to linolic acid, and the relation of an antioxidant action.

[Drawing 5] It is a figure explaining the oxidation action by ascorbic acid tetraisopulmitic acid.

[Drawing 6] It is a figure showing the outline of the experimental device used for this example.

[Drawing 7]It is a figure showing the outline of the experimental device at the time of a UV irradiation experiment.

[Description of Notations]

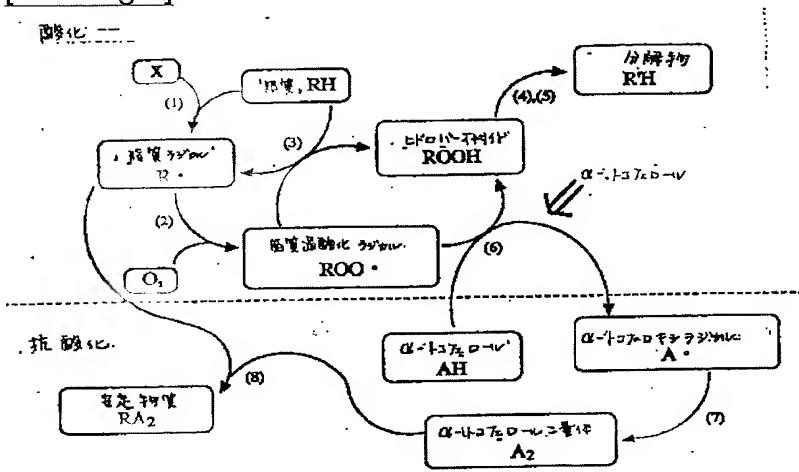
10 A reaction vessel and 12 [Opening.] A thermometer and 14 A gas loading slot and 15 A glass filter, 16 three-way cocks, and 18

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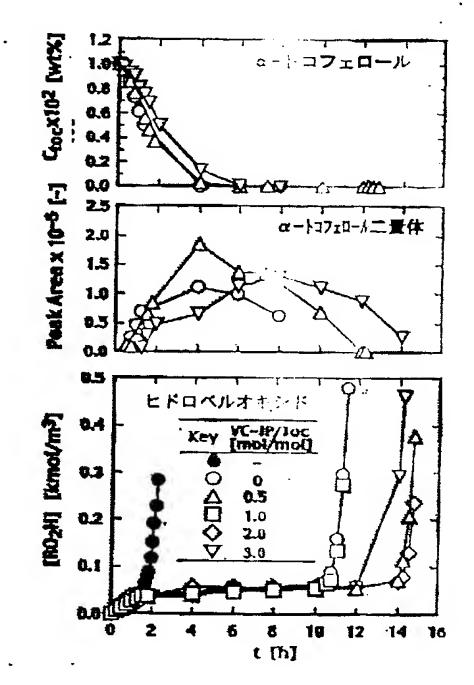
DRAWINGS

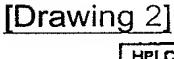
[Drawing 1]

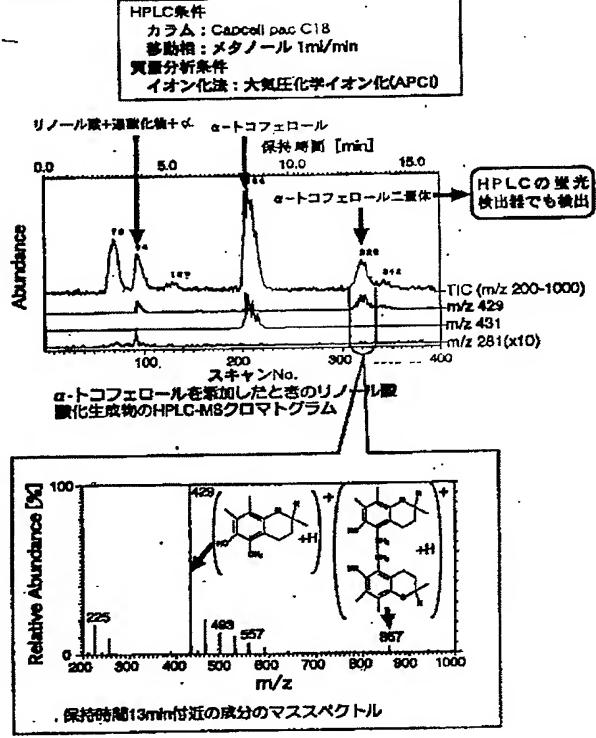


[Drawing 3]

α-トコフェロール 0.01 wi% +VC-IPの場合

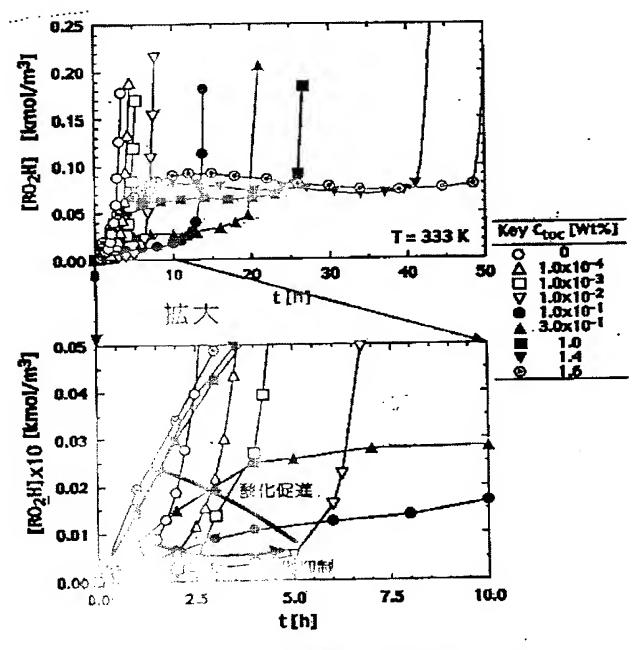






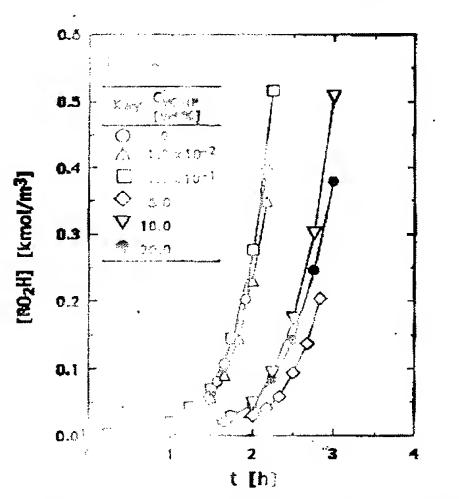
[Drawing 4]

α-トコフェロールの影響)



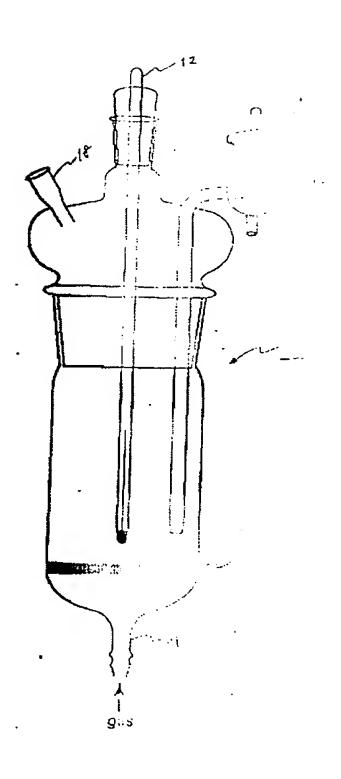
a-トコフェロールの添加量を変化させたときの リノールで、の簡化学動

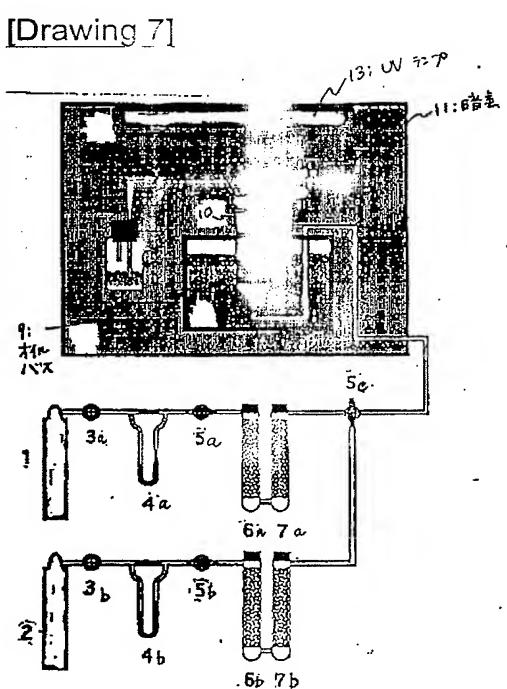
[Drawing 5]



. VC-IPの歌音は高速化させたときのリノール酸の酸化学劇

[Drawing 6]





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WRITTEN AMENDMENT

[Written amendment]

[Filing date]April 13 (2000.4.13), Heisei 12

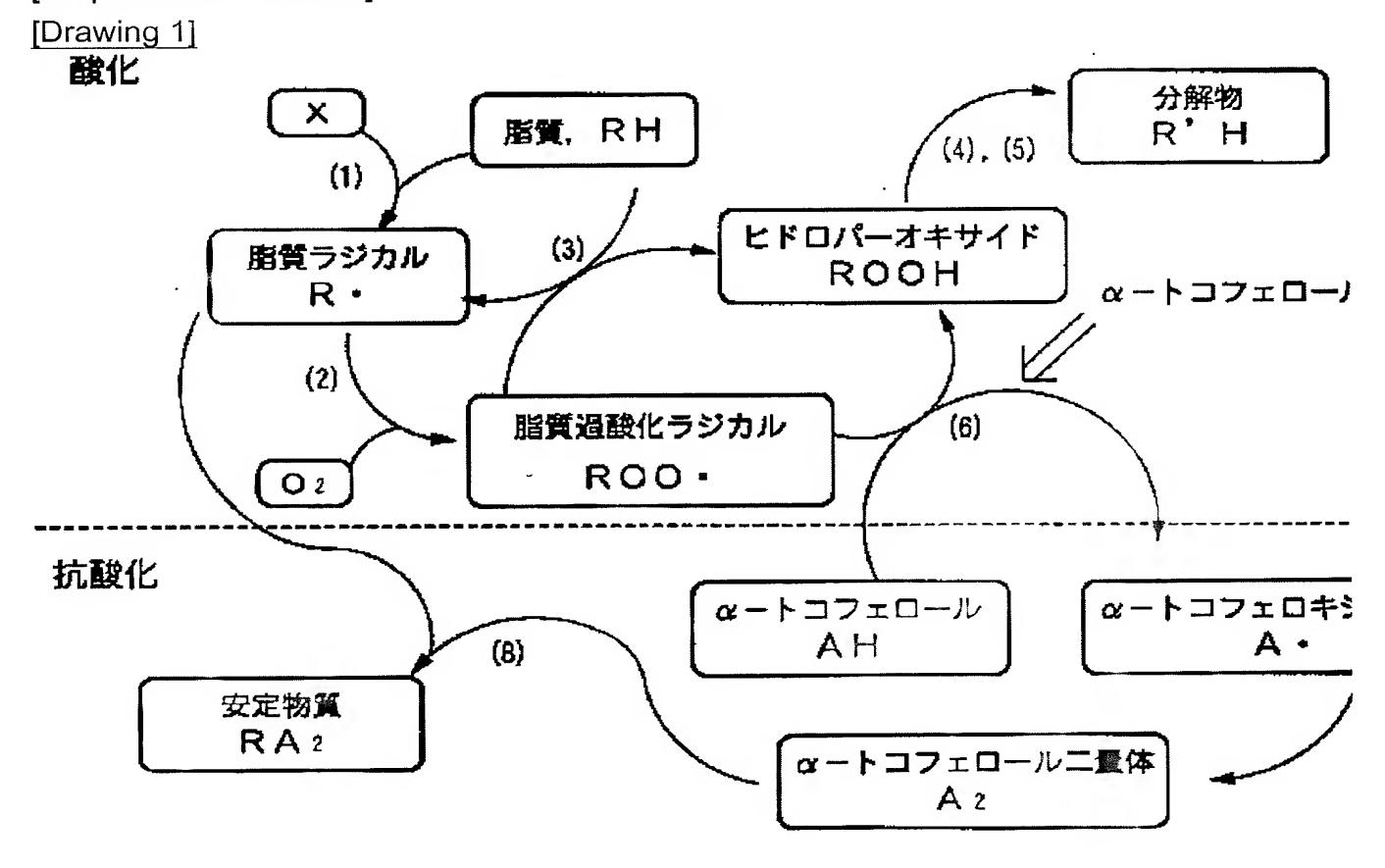
[Amendment 1]

[Document to be Amended]DRAWINGS

[Item(s) to be Amended]Drawing 1

[Method of Amendment]Change

[Proposed Amendment]



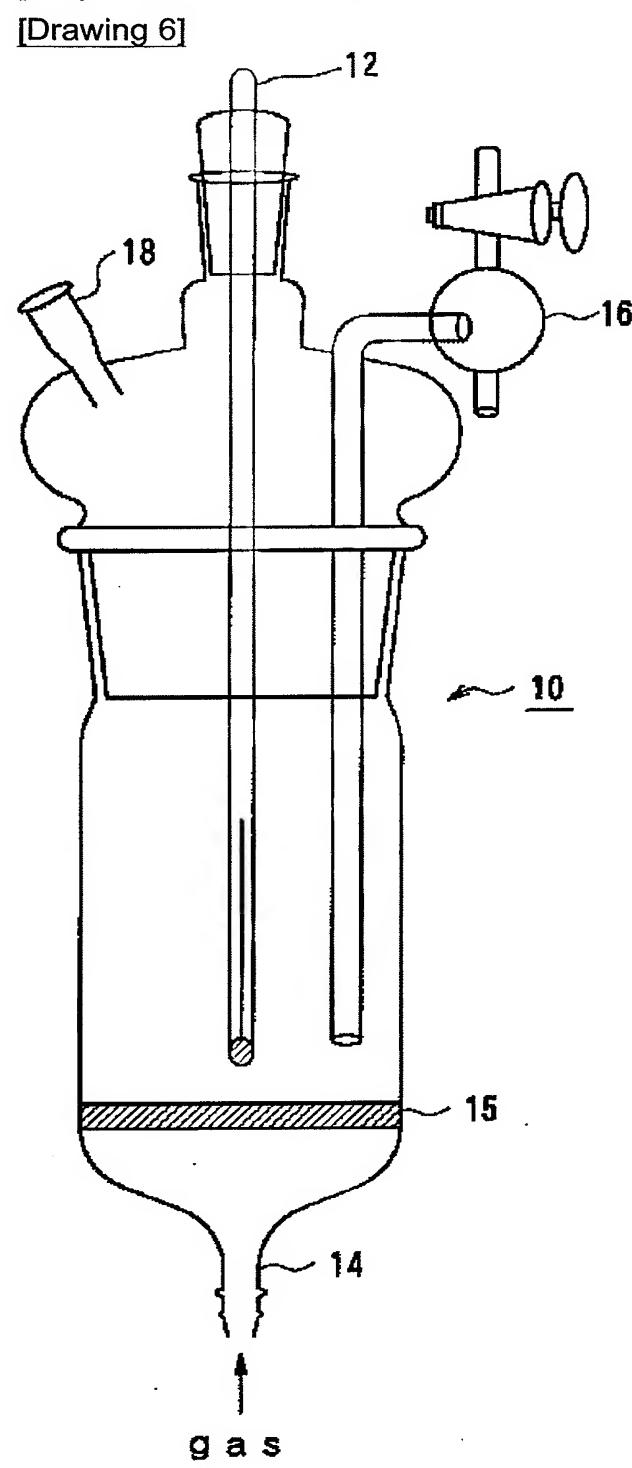
[Amendment 2]

[Document to be Amended]DRAWINGS

[Item(s) to be Amended]Drawing 6

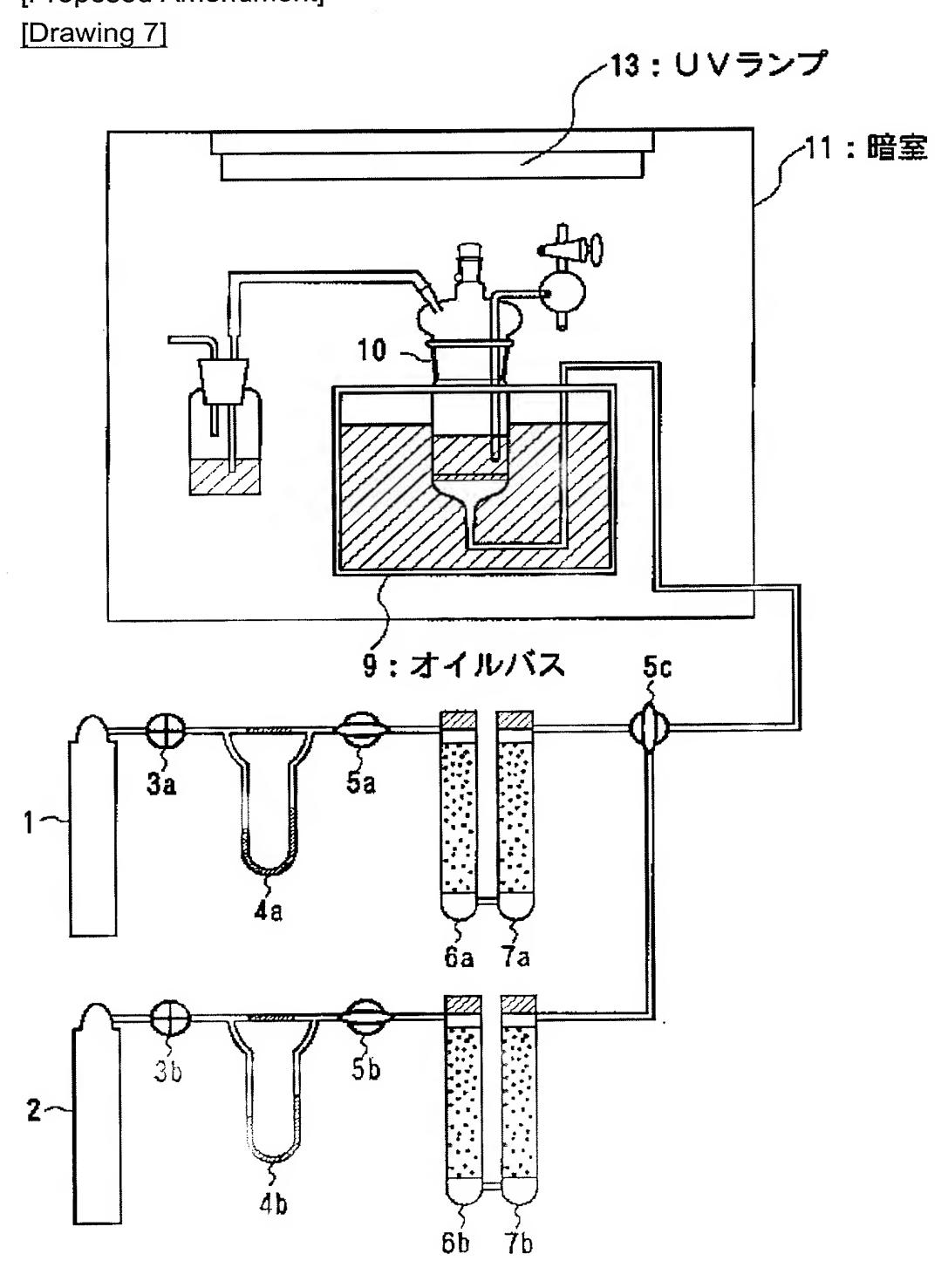
[Method of Amendment]Change

[Proposed Amendment]



[Amendment 3]
[Document to be Amended]DRAWINGS

[Item(s) to be Amended]Drawing 7 [Method of Amendment]Change [Proposed Amendment]



_____[Written amendment]

[Filing date] July 28, Heisei 12 (2000.7.28)

[Amendment 1]

[Document to be Amended]DRAWINGS

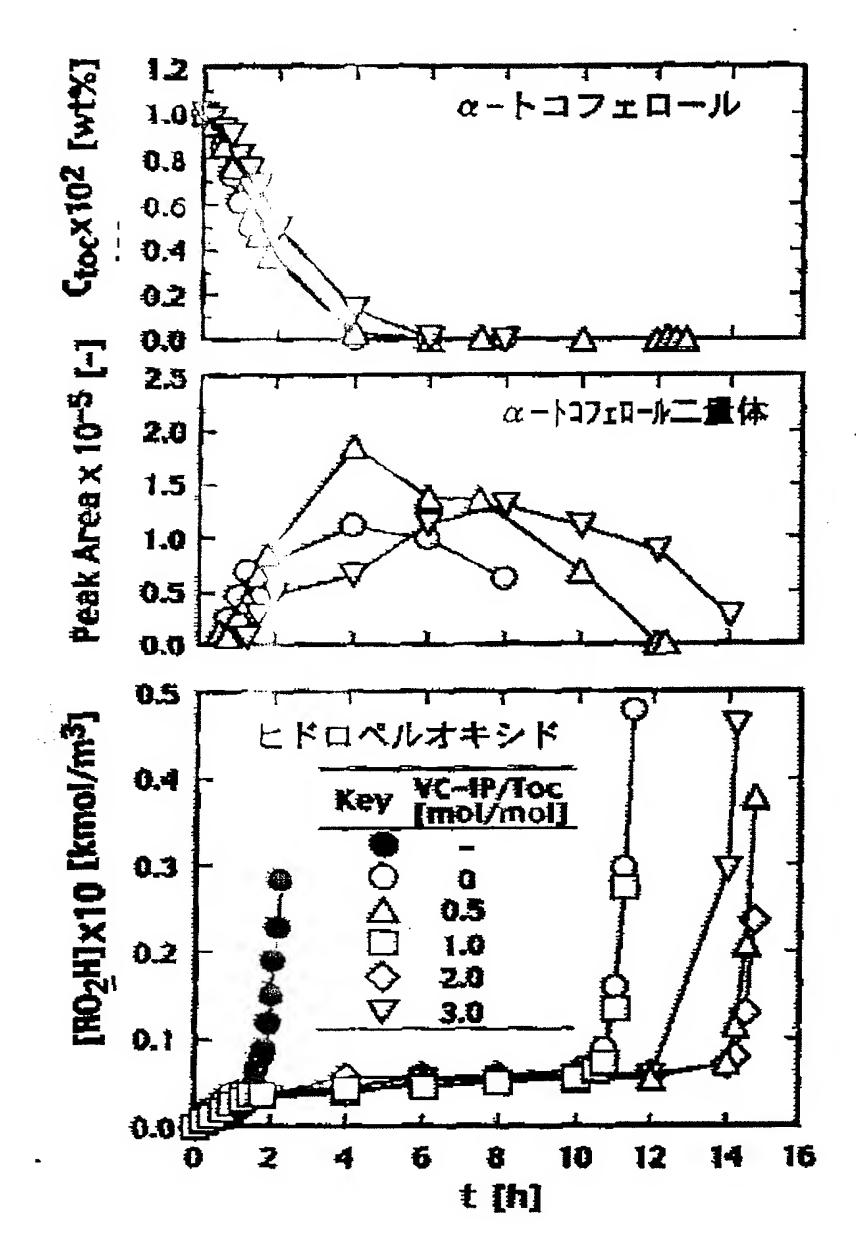
[Item(s) to be Arachded]Drawing 3

[Method of Amer. ment]Change

[Proposed Amendment]

[Drawing 3]

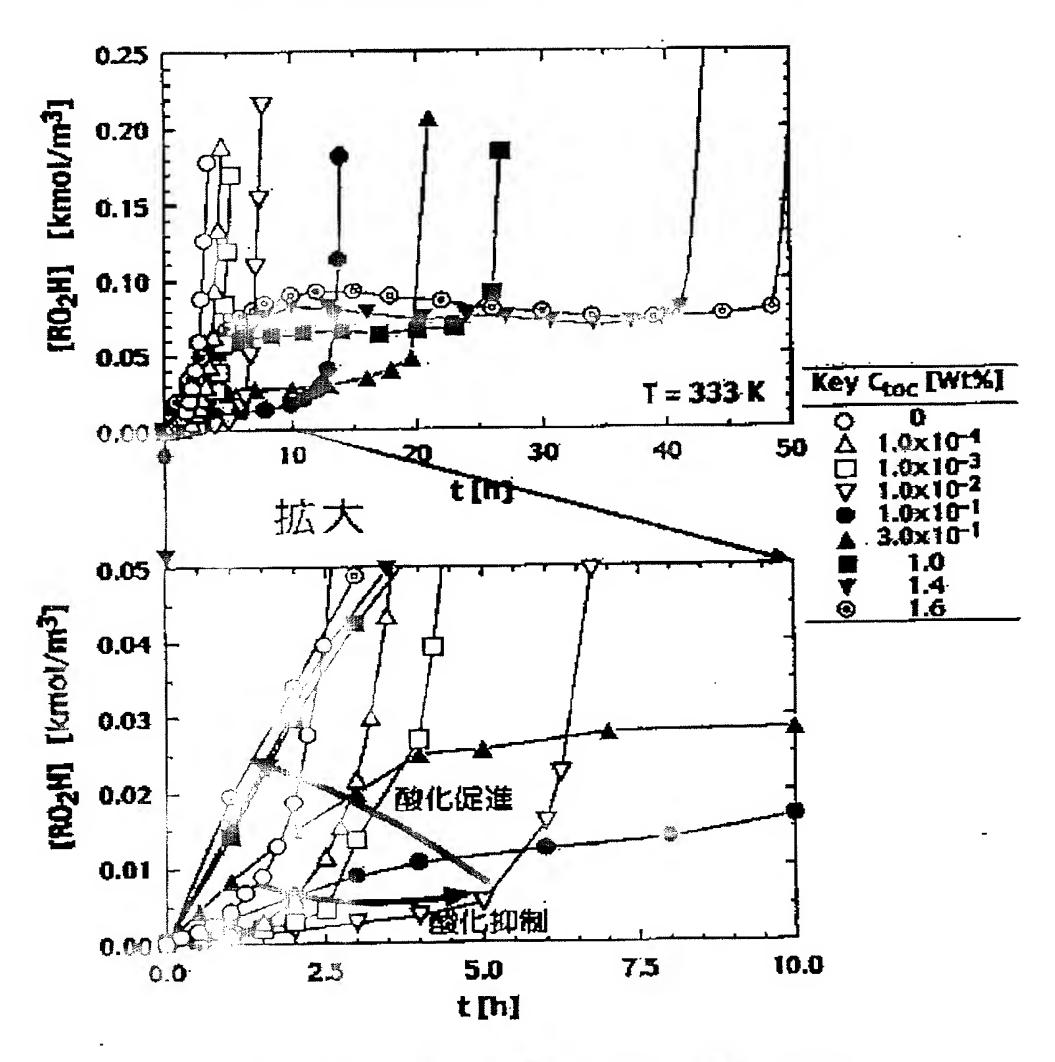
α-トコフェロール 0.01 wt% +VC-IPの場合



[Amendment 2]
[Document to be Amended]DRAWINGS
[Item(s) to be Amended]Drawing 4
[Method of Amendment]Change
[Proposed Amendment]

[Drawing 4]

α-トコフェロールの影響)



α-トコブーロールの添加量を変化させたときの リノール。&の酸化学動

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CLAIMS

[Claim(s)]

[Claim 1]An anti-oxidant containing a dimer of tocopherol.

[Claim 2]An anti-oxidant comprising:

A dimer of tocopherol.

Tocopherol.

[Claim 3]An anti-oxidant comprising:

A dimer of tocopherol.

Ascorbic acid or its derivative.

[Claim 4]An anti-oxidant comprising:

A dimer of tocopherol.

Tocopherol, and ascorbic acid or its derivative.

[Claim 5]An anti-oxidant by which a lipophilic group salt of ascorbic acid of 20 or less weight sections being included to lipid 80 weight section.

[Claim 6]Cosmetics containing the anti-oxidant according to any one of claims 1 to 5.

[Claim 7]Ultraviolet inhibitor containing the anti-oxidant according to any one of claims 1 to 5.

[Claim 8]Foodstuffs containing the anti-oxidant according to any one of claims 1 to 5.